

# DUPLICATE

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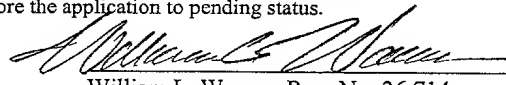
Form PTO-1390 U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE (Rev. 1-98)		Attorney's Docket Number <b>18584-0002</b>
<b>TRANSMITTAL LETTER TO THE UNITED STATES          DESIGNATED/ELECTED OFFICE (DO/EO/US)          CONCERNING A FILING UNDER 35 U.S.C. 371</b>		U.S. Application No. (if known, see 37 CFR 1.5) <b>09/937387</b>
International Application No. <b>PCT/EP00/01616</b>	International Filing Date <b>26 February 2000</b>	Priority Date Claimed <b>26 March 1999</b>
Title of Invention <b>A METHOD OF REDUCING THE DUST BEHAVIOR OF SILICATES</b>		
Applicant(s) for DO/EO/US <b>FENTEN, Norbert; WOLF, Wilfried</b>		
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:		
<ol style="list-style-type: none"> <li>1. <input checked="" type="checkbox"/> This is a FIRST submission of items concerning a filing under 35 U.S.C. 371.</li> <li>2. <input type="checkbox"/> This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371.</li> <li>3. <input checked="" type="checkbox"/> This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), and (9) and (21) indicated below.</li> <li>4. <input type="checkbox"/> The US has been elected by the expiration of 19th month from the earliest claimed priority date (Article 31).</li> <li>5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2))           <ol style="list-style-type: none"> <li>a. <input checked="" type="checkbox"/> is attached herewith (required only if not transmitted by the International Bureau).</li> <li>b. <input type="checkbox"/> has been communicated by the International Bureau.</li> <li>c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US).</li> </ol> </li> <li>6. <input checked="" type="checkbox"/> An English language translation of the International Application into English (35 U.S.C. 371(c)(2))           <ol style="list-style-type: none"> <li>a. <input checked="" type="checkbox"/> is attached herewith.</li> <li>b. <input type="checkbox"/> has been previously submitted under 35 U.S.C. 154(d)(4).</li> <li>c. <input type="checkbox"/> translation not required as the application was filed in English.</li> </ol> </li> <li>7. <input type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))           <ol style="list-style-type: none"> <li>a. <input type="checkbox"/> are attached herewith (required only if not transmitted by the International Bureau).</li> <li>b. <input type="checkbox"/> have been communicated by the International Bureau.</li> <li>c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired.</li> <li>d. <input type="checkbox"/> have not been made and will not be made.</li> </ol> </li> <li>8. <input checked="" type="checkbox"/> An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).</li> <li>9. <input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).</li> <li>10. <input type="checkbox"/> An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).</li> </ol>		
<b>Items 11. to 20. below concern document(s) or information included:</b>		
<ol style="list-style-type: none"> <li>11. <input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98.</li> <li>12. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.</li> <li>13. <input checked="" type="checkbox"/> A FIRST preliminary amendment.</li> <li>14. <input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment.</li> <li>15. <input type="checkbox"/> A substitute specification.</li> <li>16. <input type="checkbox"/> A change of power of attorney and/or address letter.</li> <li>17. <input type="checkbox"/> A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.</li> <li>18. <input type="checkbox"/> A second copy of the published international application under 35 U.S.C. 154(d)(4).</li> <li>19. <input type="checkbox"/> A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).</li> <li>20. <input type="checkbox"/> Other items or information:</li> </ol>		
Express Mail Label No. <b>EL690570512US</b>		Date: <b>September 25, 2001</b>
Page 1 of 2		

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PATENT & TRADEMARK OFFICE

U.S. Application No. <b>09/937387</b> International Application No. PCT/EP00/01616 Attorney's Docket Number 18584-0002		
21. <input checked="" type="checkbox"/> The following fees are submitted: <span style="float: right;"><u>CALCULATIONS PTO USE ONLY</u></span>		
<b>BASIC NATIONAL FEE (37 CFR 1.492(a)(1)-(5)):</b>  Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO.. <b>\$1000.00</b>  International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO..... <b>\$860.00</b>  International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO ..... <b>\$710.00</b>  International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4)..... <b>\$690.00</b>  International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4)..... <b>\$100.00</b>		
<b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b>		\$860.00
Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).		\$0.00
Claims	Number Filed	Number Extra
Total claims	12 - 20 =	0
Independent Claims	2 - 3 =	0
		Rate
		x 18.00
		x 80.00
Multiple Dependent Claims (if applicable)		+ 270.00
<b>TOTAL OF ABOVE CALCULATIONS =</b>		\$860.00
<input type="checkbox"/> Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are reduced by 1/2.		\$0.00
<b>SUBTOTAL =</b>		\$860.00
Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).		\$0.00
<b>TOTAL NATIONAL FEE =</b>		\$0.00
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property		\$0.00
<b>TOTAL FEES ENCLOSED =</b>		\$860.00
		Amount to be refunded:
		charged:
a. <input checked="" type="checkbox"/> A check in the amount of <b>\$860.00</b> to cover the above fees is enclosed.  b. <input type="checkbox"/> Please charge my Deposit Account No. 19-5029 in the amount of \$_____ to cover the above fees. A duplicate copy of this sheet is enclosed.  c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment, to Deposit Account No. 19-5029. A duplicate copy of this sheet is enclosed.		
NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.		
SEND ALL CORRESPONDENCE TO: William L. Warren, Esq. SUTHERLAND ASBILL & BRENNAN, LLP 999 Peachtree Street, N.E. Atlanta, Georgia 30309 Telephone: 404-853-8000		
 William L. Warren, Reg. No. 36,714		

PATENTS

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:	)	
	)	
<b>Norbert Fenten, et al.</b>	)	
	)	Art Unit: <b>Not yet assigned</b>
Serial No. <b>Not Yet Assigned</b>	)	
	)	Examiner: <b>Not yet assigned</b>
Filed: <b>September 25, 2001</b>	)	
	)	
For: <b>A METHOD OF REDUCING THE DUST</b>	)	
<b>BEHAVIOR OF SILICATES</b>	)	
	)	
International Application: <b>PCT/EP00/01616</b>	)	
	)	
International Filing Date: <b>February 26, 2000</b>	)	
	)	
Priority Date: <b>March 26, 1999</b>	)	

**PRELIMINARY AMENDMENT**

Box New Patent Application  
Commissioner for Patents  
Washington, DC 20231

Dear Sirs:

Please enter the following amendments prior to the first evaluation of this application.

**AMENDMENTS**

Please cancel Claims 1-12 without prejudice or disclaimer. Please enter the following new Claims 13-24.

13. A method of reducing the dust behavior of silicates, wherein the comminuted silicate is surface-coated with a polysiloxane-polyether copolymer.
14. The method according to claim 13, wherein from 0.1 to 4 wt.-% of polysiloxane-polyether copolymer is used in coating.

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15. The method according to claim 13, wherein from 0.5 to 1.5 wt.-% of polysiloxane-polyether copolymer is used in coating.
16. The method according to claim 13, wherein calcium metasilicate is used as the silicate.
17. The method according to claim 13, wherein wollastonite is used as the silicate.
18. A silicate having reduced dust behavior, wherein said silicate has been surface-coated with polysiloxane-polyether copolymer and exhibits the following properties:

Specific surface area (DIN66132-BET)	0.5 to 1.5m <sup>2</sup> /g,
Dust content (according to Heubach)	500-1200 mg/100g.
19. The silicate according to claim 18, wherein said silicate is a calcium metasilicate.
20. The silicate according to claim 18, wherein said silicate is wollastonite.
21. The silicate according to claim 18, wherein said silicate has been surface-coated with 0.1 to 4 wt.-% of polysiloxane-polyether copolymer.
22. The silicate according to claim 18, wherein said silicate has been surface-coated with 0.5 to 1.5 wt.-% of polysiloxane-polyether copolymer.
23. Use of the silicate according to claim 18 as a reinforcing filler in plastic materials.
24. The use according to claim 23 as a reinforcing filler in plastic materials selected from the group of polyurethane, polyamide, and polypropylene.

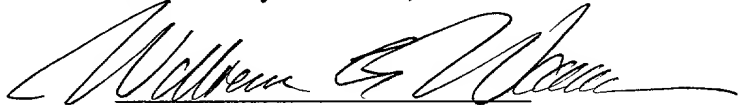
**REMARKS**

Applicants have cancelled claims 1-12 as were pending due to substitution in the Response to the Written Opinion in the PCT stage of this application. Applicants submit new claims 13-24 herein, and on separate clean sheets, which contain no new matter. Applicants believe the new claims are directed to allowable subject matter.

The filing fee for this application as calculated in PTO-1390 attached considers the claim amendments presented in this Preliminary Amendment.

The Examiner is encouraged to call the undersigned attorney if doing so will facilitate prosecution of the application. No additional fees are believed due, however, the Commissioner is hereby authorized to charge any fees due or credit any overpayment to Deposit Account 19-5029.

Respectfully submitted,



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Docket No.: 18584-0002

**CLAIMS PENDING AFTER AMENDMENT HEREIN**

13. A method of reducing the dust behavior of silicates, wherein the comminuted silicate is surface-coated with a polysiloxane-polyether copolymer.
14. The method according to claim 13, wherein from 0.1 to 4 wt.-% of polysiloxane-polyether copolymer is used in coating.
15. The method according to claim 13, wherein from 0.5 to 1.5 wt.-% of polysiloxane-polyether copolymer is used in coating.
16. The method according to claim 13, wherein calcium metasilicate is used as the silicate.
17. The method according to claim 13, wherein wollastonite is used as the silicate.
18. A silicate having reduced dust behavior, wherein said silicate has been surface-coated with polysiloxane-polyether copolymer and exhibits the following properties:
- |                                      |                              |
|--------------------------------------|------------------------------|
| Specific surface area (DIN66132-BET) | 0.5 to 1.5m <sup>2</sup> /g, |
| Dust content (according to Heubach)  | 500-1200 mg/100g.            |
19. The silicate according to claim 18, wherein said silicate is a calcium metasilicate.
20. The silicate according to claim 18, wherein said silicate is wollastonite.
21. The silicate according to claim 18, wherein said silicate has been surface-coated with 0.1 to 4 wt.-% of polysiloxane-polyether copolymer.
22. The silicate according to claim 18, wherein said silicate has been surface-coated with 0.5 to 1.5 wt.-% of polysiloxane-polyether copolymer.

23. Use of the silicate according to claim 18 as a reinforcing filler in plastic materials.
24. The use according to claim 23 as a reinforcing filler in plastic materials selected from the group of polyurethane, polyamide, and polypropylene.

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**Claims:**

- 5 1. A method of reducing the dust behavior of silicates, wherein the com-  
minuted silicate is surface-coated with a polysiloxane-polyether copoly-  
mer.
- 10 2. The method according to claim 1, characterized in that from 0.1 to  
4 wt.-% of polysiloxane-polyether copolymer is used in coating.
3. The method according to claim 1 or 2, characterized in that from 0.5 to  
1.5 wt.-% of polysiloxane-polyether copolymer is used in coating.
- 15 4. The method according to claims 1 to 3, characterized in that calcium me-  
tasilicate is used as silicate.
5. The method according to claims 1 to 4, characterized in that wollastonite  
is used as silicate.
- 20 6. A silicate having reduced dust behavior, characterized in that said silicate  
has been surface-coated with polysiloxane-polyether copolymer and ex-  
hibits the following properties:  
Specific surface area (DIN66132-BET) 0.5 to 1.5 m<sup>2</sup>/g,  
25 Dust content (according to Heubach) 500-1200 mg/100g.
7. The silicate according to claim 6, characterized in that said silicate is a  
calcium metasilicate.
- 30 8. The silicate according to claim 6 or 7, characterized in that said silicate is  
wollastonite.
9. The silicate according to claims 6 to 8, characterized in that said silicate  
has been surface-coated with 0.1 to 4 wt.-% of polysiloxane-polyether  
35 copolymer.



10. The silicate according to claims 6 to 9, characterized in that said silicate has been surface-coated with 0.5 to 1.5 wt.-% of polysiloxane-polyether copolymer.
- 5 11. Use of the silicate according to claims 6 to 10 as a reinforcing filler in plastic materials.
12. The use according to claim 11 as a reinforcing filler in plastic materials selected from the group of polyurethane, polyamide, and polypropylene.

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## A Method of Reducing the Dust Behavior of Silicates

The invention is directed to a method of reducing the dust behavior of silicates. The invention is also directed to a silicate having reduced dust behavior.

Silicates, particularly needle-shaped silicates such as wollastonite, which is a chain silicate, are being employed as reinforcing fillers in plastic uses.

Wollastonite is a natural calcium silicate of formula  $\text{Ca}_3[\text{Si}_3\text{O}_9]$ . Monoclinic wollastonite is comprised of  $\text{SiO}_4$  tetrahedrons linked in a three-membered arrangement of formula  $[\text{Si}_3\text{O}_9]^{6-}$  wherein the individual chains are linked via  $\text{Ca}^{2+}$  ions. This explains why wollastonite particles may have a needle-shaped structure.

In addition, wollastonite has beneficial material properties, including the absence of chemically bound water which in other hydrous silicates gives rise to changes in structure by loss of water upon heating. When incorporated in plastic materials, the result would be that a reinforcing effect no longer could be achieved. Further advantages are a low degree of shrinkage upon drying and heating, an exceedingly low expansion coefficient, and high mechanical properties of manufactured plastic products including wollastonite. In addition to its use as an additive in plastic materials, wollastonite is also used in the manufacture of ceramic products, sanitary articles, and porcelain.

The present invention predominantly relates to the use of wollastonite in plastic materials.

In such uses, wollastonite as a filling material is produced by grinding recovered wollastonite, followed by air

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classification. As a result, needle-shaped wollastonite particles are obtained, which either are incorporated directly in the plastic mixture or incorporated in the plastic materials in the form of polymer masterbatches including levels of from 10 to 40 wt.-% of wollastonite. Owing to the needle-shape structure of wollastonite, a substantial improvement in the mechanical properties is achieved in the plastic material and plastic molded articles produced therefrom.

In addition, according to the prior art, wollastonite frequently is coated with silane compounds in order to achieve improved incorporation in the plastic material. To this end, surface coating using aminosilane, epoxysilane, methacrylic silane, trimethylsilane, vinylsilane, or alkylsilane is performed.

However, these prior art silicate materials involve the disadvantage of exhibiting increased dust formation due to an increased dispersity as a result of silane coating. The dust portion is comprised of most finely particulate silicate particles essentially lacking the needle-shape structure and therefore being incapable of inducing any reinforcing effects in the plastic material. That is, part of the silicate filler is not effective, acting merely as a non-reinforcing powdered filling material.

Initially, attempts have been made to overcome this drawback by subjecting wollastonite to various procedures in order to decrease the dust behavior. However, such procedures are relatively costly and expensive, thereby resulting in a substantial rise in cost of this raw material, rendering it useless as reinforcing filler in plastic materials for cost reasons. Therefore, this raw material cannot compete with other raw materials such as fiber glass or other fibrous materials also used as reinforcing fillers.

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It was therefore the technical object of the invention to provide a simple method of reducing the dust behavior of silicates, particularly of wollastonite, which method would be practicable at a cost as low as possible and result in a substantial decrease in the dust behavior of wollastonite.

Said technical object is accomplished by subjecting the comminuted silicate, particularly wollastonite, to a surface coating using a polyorganosiloxane.

In contrast to previous surface coating using silanes, coating using polyorganosiloxanes was found to result in a substantial reduction of the dust behavior of much more than 50% as compared to prior art materials. Surface coating of silicates is a simple, cost-effective process. Thus, the appropriate surface coating agents normally are coated onto the surface by mixing the material in a fluid mixer. Furthermore, this technique has become well-tried in coating using appropriate silane compounds. Owing to this simple method of reducing the dust behavior of silicates, the previous multi-stage physical processes of reducing the dust behavior are no longer necessary and can be replaced in a simple fashion.

In particular, it has been surprising that the dust behavior in surface coating using appropriate silane compounds is not reduced or only to a minor degree, while coating using polyorganosiloxanes results in a significant reduction of the dust behavior. In this context, adequate comparative experiments were carried out which will be described in the experimental section of this application.

In a particularly preferred embodiment, a polyorganosiloxane in an amount of from 0.1 to 4 wt.-%, more preferably from 0.5 to 1.5 wt.-% is used in surface coating. A poly-

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siloxane-polyether copolymer is employed as a particularly preferred polyorganosiloxane.

As has been mentioned above, these polyorganosiloxanes allow for a reduction of the dust behavior of silicates, particularly of calcium metasilicates, and in a particularly preferred fashion, of wollastonites.

Surprisingly, this dust-reducing surface coating was found to achieve good incorporation of the filler in the plastic material. By using the polyorganosiloxane compounds of the invention, the same effect is achieved as with the corresponding functional silane compounds known from prior art.

The invention is also directed to a silicate having reduced dust behavior, which silicate is surface-coated with a polyorganosiloxane, has a Heubach dust content of from 500 to 1200 mg/100 g, the silicate having a specific surface area of from 0.5 to 1.5 m<sup>2</sup>/g, as measured according to DIN 66132-BET.

The silicate preferably is a calcium metasilicate, and more preferably a wollastonite. The silicate preferably is surface-coated using from 0.1 to 4 wt.-%, more preferably from 0.5 to 1.5 wt.-% of polyorganosiloxane. A polysiloxane-polyether copolymer preferably is employed as polyorganosiloxane. The silicate is used as a reinforcing filler in plastics, particularly in polyurethane, polyamide, and polypropylene. The silicate is produced by grinding the wollastonite on an appropriate size reduction unit, such as a counterflow impact grinding unit. The resulting comminuted product then is removed from the size reduction unit and surface coated with the polyorganosiloxane in a fluid mixer. Coating is effected at temperatures between 40 and 100°C resulting from frictional heat. Following coating, the material is cooled to room temperature. A low-dust coated

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wollastonite having the above-mentioned properties is obtained as final product.

The product according to the invention will be characterized in more detail in the Figures which follow.

Fig. 1 shows an FT-IR spectrum of the obtained product as a pressed pellet. The Figure shows the transmittance in % as a function of the wave number in  $\text{cm}^{-1}$ .

Fig. 2 shows another FT-IR spectrum of the obtained product. Assessment is effected as an illustration of the absorbance as a function of the wave number in  $\text{cm}^{-1}$ . The wollastonite raw material has been subtracted mathematically from the FT-IR spectrum.

Fig. 3 shows the weight ratio of the product of the invention as a function of the needle length. Therein, the needle length is given in  $\mu\text{m}$  needle length classifications.

Fig. 4 shows the weight ratio of the product of the invention as a function of the needle diameter. Therein, the needle diameter is given in  $\mu\text{m}$  needle diameter classifications.

Ultimately, Fig. 5 shows the length/diameter average ratio of the product of the invention as a function of needle length. The needle length is given in  $\mu\text{m}$  needle length classifications.

In a preferred fashion, this material is employed as a reinforcing filler in plastic materials, resulting in an improvement of the mechanical properties, particularly the tensile strength and reinforcement of the composite material. In a particularly preferred fashion, the use as a reinforcing filler is in plastic materials selected from the

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group of polyurethane, polyamide, polypropylene. Further advantages are to be seen in a high throughput capacity on extruders owing to good dispersibility and high thermal conductivity of the material, low shrinkage and high dimensional stability of the wollastonite-containing polymer masterbatch resulting from a reduction of the linear expansion coefficient. Plastic articles produced using this material as reinforcing filler have excellent surface quality owing to the finely particulate needle structure of the material. Furthermore, heat distortion resistance is improved and the creep tendency is reduced. By using the reinforcing fillers, an increase of the surface hardness of the masterbatch is achieved as a result of the hardness of the filler, with optimum incorporation of the material in the plastic.

Without intending to be limiting, the invention will be illustrated in more detail with reference to the Examples below.

## **Examples**

### **Example 1**

#### **Production of polyorganosiloxane-coated wollastonite**

1000 kg of wollastonite is comminuted on a counterflow impact grinding unit (Majac). The jet mill operates with 2 boron carbide nozzles at an operating pressure of 7 bars. The classifier used to separate non-comminuted coarse particles is operated at a rotational speed of 1800 rpm. Separation of the final product is effected in a filter.

75 kg of wollastonite thus comminuted is coated in a Thyssen-Henschel FM250D fluid mixer using 750 g of a

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polysiloxane-polyether copolymer (TEGOSTAB B 8427, TH. GOLDSCHMIDT AG, Essen, Germany).

The rotational speed of the mixing tools (bottom scraper, fluidizing blades, horn tools) is 1200 rpm. The fluid mixer is double-walled and enables adjusting particular temperature patterns independent of the frictional heat by indirect heating with hot water or steam. The temperature pattern adjusted in the wollastonite reaches 80°C.

Thereafter, the coated wollastonite having a material temperature of 80°C is cooled to room temperature in a double-walled cooling mixer (Thyssen-Henschel Type 650). The mixing tools are comparable to the mixing tools of the fluid mixer. The rotational speed is 140 rpm.

A low-dust, coated wollastonite is obtained as final product.

Fig. 1 shows an FT-IR spectrum of the obtained product as a pressed pellet. The Figure shows the transmittance in % as a function of the wave number in  $\text{cm}^{-1}$ . The vibrational bands are characteristic for the polyorganosiloxane-coated wollastonite system.

Fig. 2 shows another FT-IR spectrum of the obtained product. Assessment is effected as an illustration of the absorbance as a function of the wave number in  $\text{cm}^{-1}$ . Only wave numbers of from 2,400 to 3,600 have been covered, and the wollastonite raw material has been subtracted mathematically from the FT-IR spectrum within the scope of the present analysis. As a consequence, Figure 2 merely shows the vibrational bands of the surface coating, namely, of the polyorganosiloxane on the surface of wollastonite, particularly in the wave number range of from 2,800 to 3,100. In particular, the CH vibrations are typical in this wave num-



ber range. Therefore, the illustration thus obtained is a characteristic fingerprint of the surface additive, describing the specific structure of the polyorganosiloxane employed.

Figures 3, 4 and 5 characterize the obtained product with respect to needle length (Figure 3), needle diameter (Figure 4), and average L/D ratio (length/diameter), as a function of the needle length (Figure 5).

## **Example 2**

### **Dust measurement**

In this Example, the product of the invention produced according to Example 1 was examined for its dust behavior in comparison to prior art products. More specifically, the following products were tested:

- a) wollastonite according to Example 1;
- b) wollastonite with no additive (prior art);
- c) wollastonite with aminosilane as additive (prior art).

The dust behavior was determined using the following method:

1. Dust measurement using the Heubach Dustmeter
2. Determination of the specific surface according to DIN 66132-BET

#### *1. Dust measurement using the Heubach Dustmeter*

A Heubach Dustmeter, an analytical balance, as well as a filter GF 92 (S+S) are used in this investigation. The measuring instrument consists of a base instrument with air

- holders for mounted structures
- dust formation vessel
- coarse separator
- air filtration vessel

The suction pipe with tube connector is screwed tightly on the threaded socket, the stabilizer pipe is fitted onto the bolt, fixed with the clamping block, and the foot is secured to the pipe ends. The short stand rods can be fixed at each position of the bracket. The desired array is furnished using the double sleeve and the stand clamp.

The substance to be tested is kept in motion in the rotating dust formation vessel. The dust is taken up by an axially entering stream of air and is moved past a coarse separator made of glass wherein coarse, non-dispersible particles undergo sedimentation. The dust remaining in the stream of air is deposited on a filter. The air pressure required in testing is generated by a vacuum pressure pump, the air meter measurement being effected on the pressure side.

Adjusting the desired air flow:

One unit of the counter corresponds to 0.1 l, i.e., for e.g. 100 l of air, "1000" must be preselected on the set-point generator of the four-digit counter. The air filtration vessel covered with filter paper is connected to the pump, the needle valve on the flowmeter is fully opened,

d) Dust measurement

Prior to beginning the measurement, the air filtration vessel covered with filter paper is weighed ( $m_1$ ). The desired amount of air is preselected on the four-digit digital set-point generator. Once all the connections from the dust formation vessel up to the pump are established, the "START" button is pressed, setting both the gear motor and the pump in operation. Once the preselected amount of air is reached, both of them will be stopped automatically. Reweighing of the filtration vessel furnishes  $m_2$ . Prior to weighing, the rubber plug remaining on the filter should be cleansed on its outside using a cleaning paper.

In any case, air flowing through the measuring instrument is passed through an upstream drying tower in order to eliminate the effect of humidity on dust formation.

Air volume: 20 l/min  
Measuring period: 5 min  
Initial weight: 50 g  
Filter: GF 92, Schleicher & Schüll

f) Assessment

The dust formation number "s" corresponds to the amount of dust in mg formed from 100 g of test material under standardized conditions.

Said number is calculated according to the following equation:

$$s = \frac{m_2 - m_1}{m_0} \times 10^5$$

Therein:

- $m_0$  represents the initial weight of sample in g;  
 $m_1$  represents the mass of the filter casing with inserted filter prior to measurement;  
 $m_2$  represents the mass of the filter casing with inserted filter after measurement.

The dust formation number "s" of all of the three samples was determined. Table 1 shows the dust formation number in mg/100 g.

Table 1

Material	Dust formation number [mg/100 g]
Wollastonite of Example 1	700
Wollastonite with no additive	1200
Wollastonite with aminosilane	1500

As can be seen from Table 1, the wollastonite according to Example 1 has a dust formation number which is by 50% lower than that of prior art wollastonite coated with aminosilane.

2. *Determination of the specific surface area according to DIN 66132-BET*

In the following test, the specific surface area of the wollastonite according to Example 1 and of prior art comparative products was determined. The specific surface area is particularly high in materials having a high dust content, because dust has a large specific surface area. For these products, the following data were determined, which are illustrated in Table 2.

**Table 2**

Material	Specific surface area [m <sup>2</sup> /g]
Wollastonite of Example 1	0.8
Wollastonite with aminosilane	1.2

As can be seen from Table 2, the specific surface area of the product according to the invention is lower than the specific surface area of wollastonite with aminosilane.

**Example 3**

**Production of a plastic (polyurethane) molded article including reinforcing wollastonite filler**

Using the wollastonite product of Example 1 and a comparative product, namely, wollastonite coated with aminosilane, a plastic molded article having the following composition was produced:

Polyol

Polyether 64.4 parts by weight glycerol/85  
PO/15 EO, OH number = 35  
Diethyltoluylenediamine 35 parts by weight  
Diazabicyclooctane 0.5 parts by weight  
Dibutyltin dilaurate 0.1 parts by weight

Semi-prepolymer

Polyether glycerol/85 PO/15 EO/OH number = 35  
Diphenylmethane-4,4'-diisocyanate (NCO content 18%)

The mixing ratio was 100 parts by weight of polyol and 107 parts by weight of semi-prepolymer, as well as 20 parts by weight of wollastonite in the elastomer. Using the plastic material thus produced, a plastic molded article was injection-molded and examined for its mechanical properties. As illustrated in Table 4, the following results were obtained:

Table 4

	Wollastonite with aminosilane	Wollastonite of Example 1
Shore D hardness (DIN 53505)	63	64
Tensile strength (DIN 53455)	25	25
Elongation at break, % (DIN 53455)	100	130
Modulus in flexure, MPa (DIN 53457)	1600	1800
Shrinkage, % (DIN 53464)	0.5	0.4

As can be seen from the Table, the mechanical properties of the plastic material wherein the wollastonite of Example 1 is used are superior over the mechanical properties of conventionally reinforced plastic materials.

#### Example 4

#### Production of a plastic (polypropylene) molded article including reinforcing wollastonite filler

A polypropylene copolymer is produced with a level of 20 wt.-% of wollastonite of Example 1 and with a comparative product, namely, wollastonite coated with alkylsilane. Using this material, a plastic molded article plastic is produced and its mechanical properties are determined. As illustrated in Table 5, the following results were obtained:

Table 5

	Wollastonite with alkylsilane	Wollastonite of Example 1
Tensile strength, MPa (DIN 53455)	18.4	19.1
Elongation at break, % (DIN 53455)	30.4	17.9
Modulus in flexure, GPa (DIN 53457)	2.266	2.233
Impact strength (Izod) (kJ/m <sup>2</sup> )		
unnotched, Iso 180	50.4	89.0
notched, Iso 180	19.0	23.5
Heat distortion temperature HDT-B (°C) DIN 53461	123	115

As can be seen from the Table, the mechanical properties of the plastic material wherein the wollastonite of Example 1 is used are improved over conventionally reinforced polypropylene.

**Claims:**

1. A method of reducing the dust behavior of silicates, characterized in that the comminuted silicate is surface-coated with a polysiloxane-polyether copolymer.
2. The method according to claim 1, characterized in that from 0.1 to 4 wt.-% of polysiloxane-polyether copolymer is used in coating.
3. The method according to claim 1 or 2, characterized in that from 0.5 to 1.5 wt.-% of polysiloxane-polyether copolymer is used in coating.
4. The method according to claims 1 to 3, characterized in that calcium metasilicate is used as silicate.
5. The method according to claims 1 to 4, characterized in that wollastonite is used as silicate.
6. A silicate having reduced dust behavior, characterized in that said silicate has been surface-coated with polysiloxane-polyether copolymer and exhibits the following properties:  
Specific surface area (DIN66132-BET) 0.5 to 1.5 m<sup>2</sup>/g,  
Dust content (according to Heubach) 500-1200 mg/100g.
7. The silicate according to claim 6, characterized in that said silicate is a calcium metasilicate.
8. The silicate according to claim 6 or 7, characterized in that said silicate is wollastonite.

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105260 1824360



9. The silicate according to claims 6 to 8, characterized in that said silicate has been surface-coated with 0.1 to 4 wt.-% of polysiloxane-polyether copolymer.
10. The silicate according to claims 6 to 9, characterized in that said silicate has been surface-coated with 0.5 to 1.5 wt.-% of polysiloxane-polyether copolymer.
11. Use of the silicate according to claims 6 to 10 as a reinforcing filler in plastic materials.
12. The use according to claim 11 as a reinforcing filler in plastic materials selected from the group of polyurethane, polyamide, and polypropylene.

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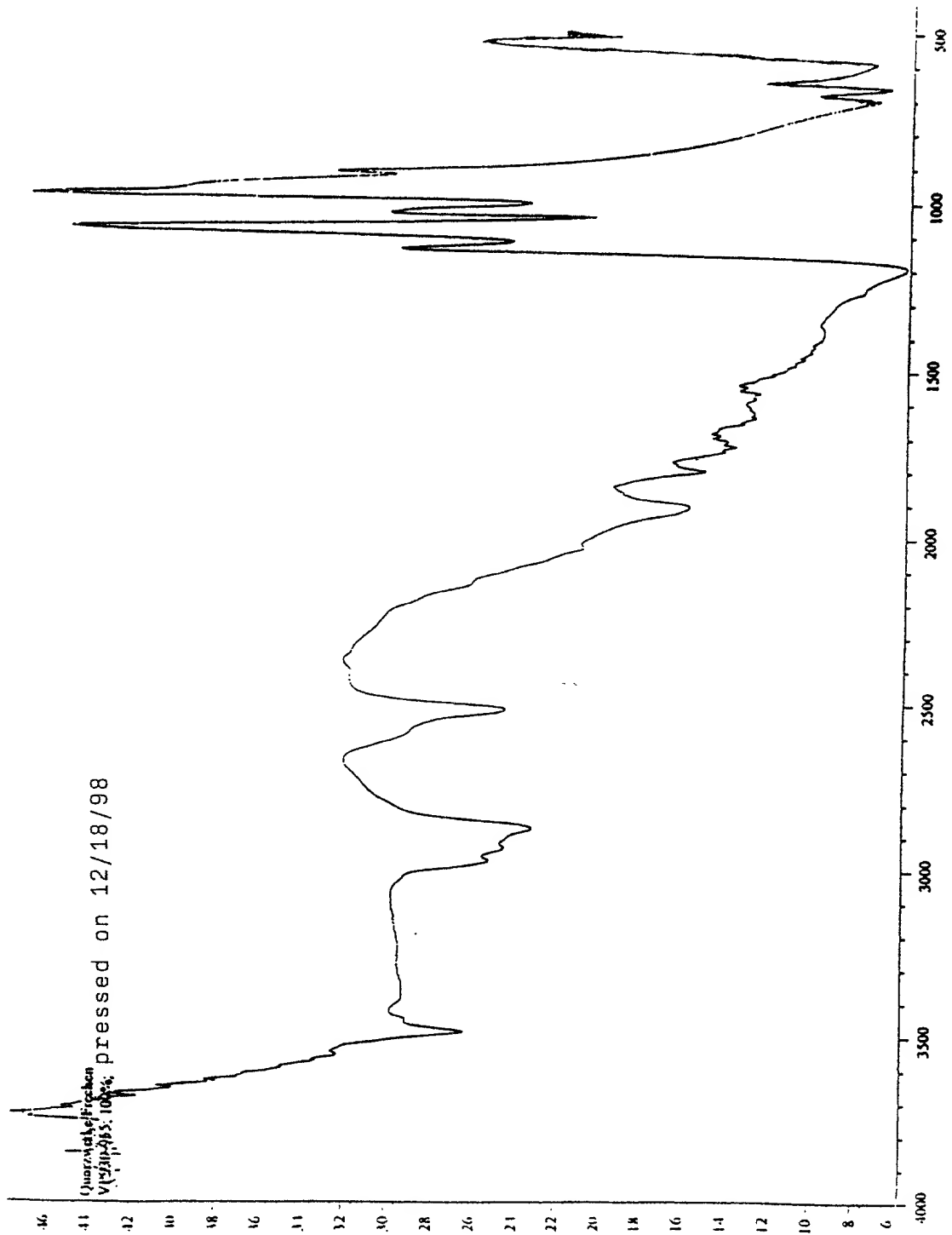
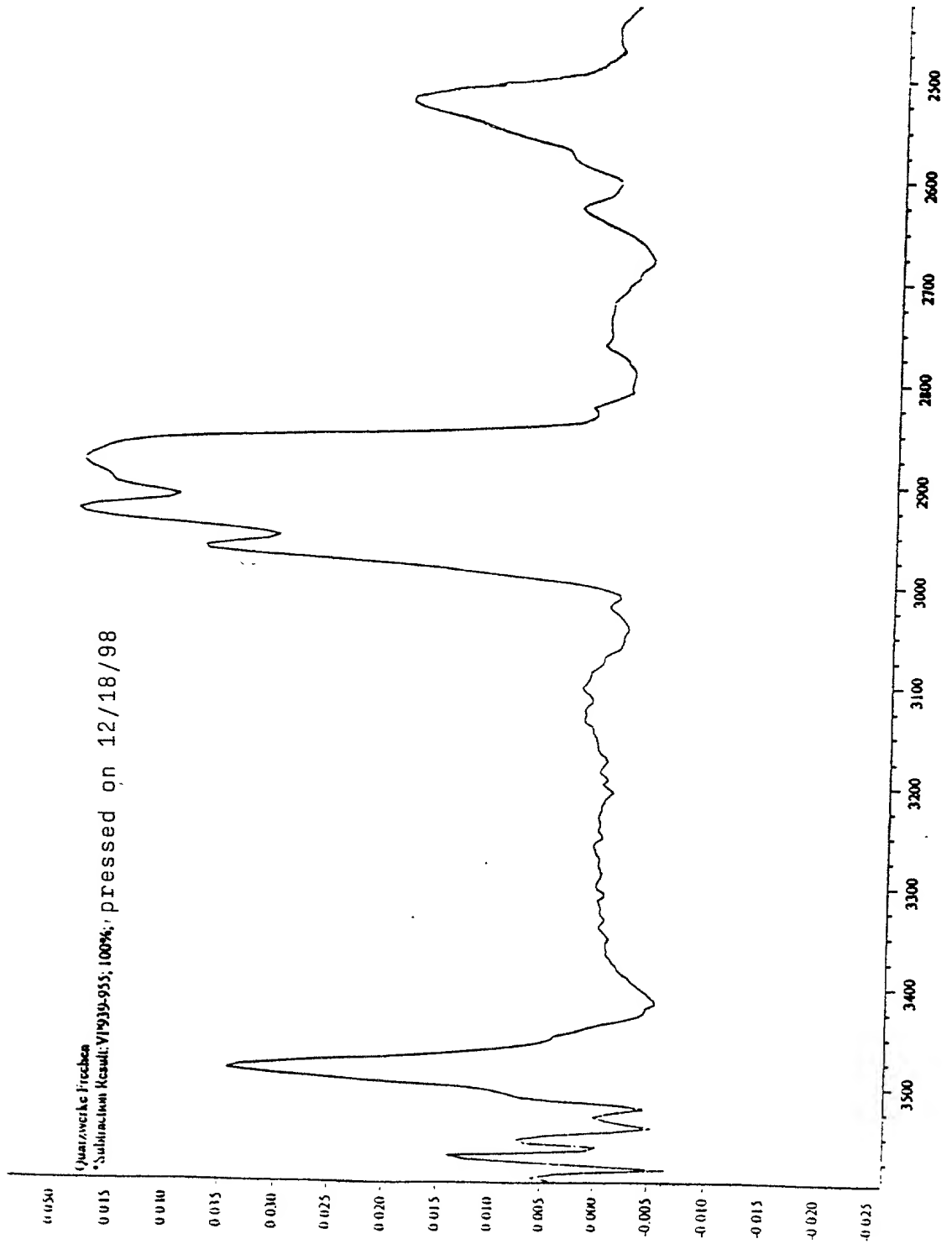


Figure 1

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Figure 2



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Figure 3

Weight ratio as a function of needle length

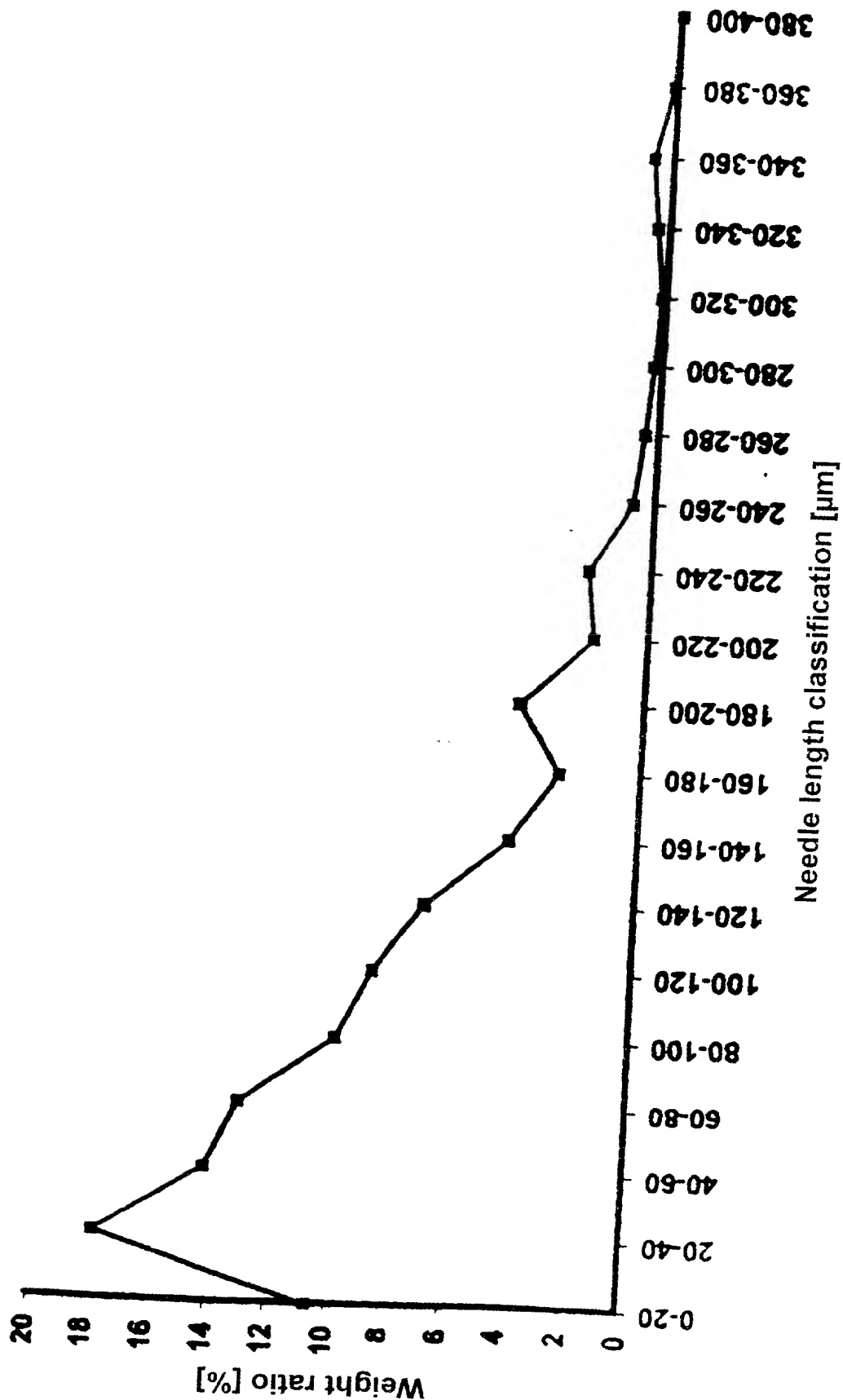


Figure 4

Weight ratio as a function of needle diameter

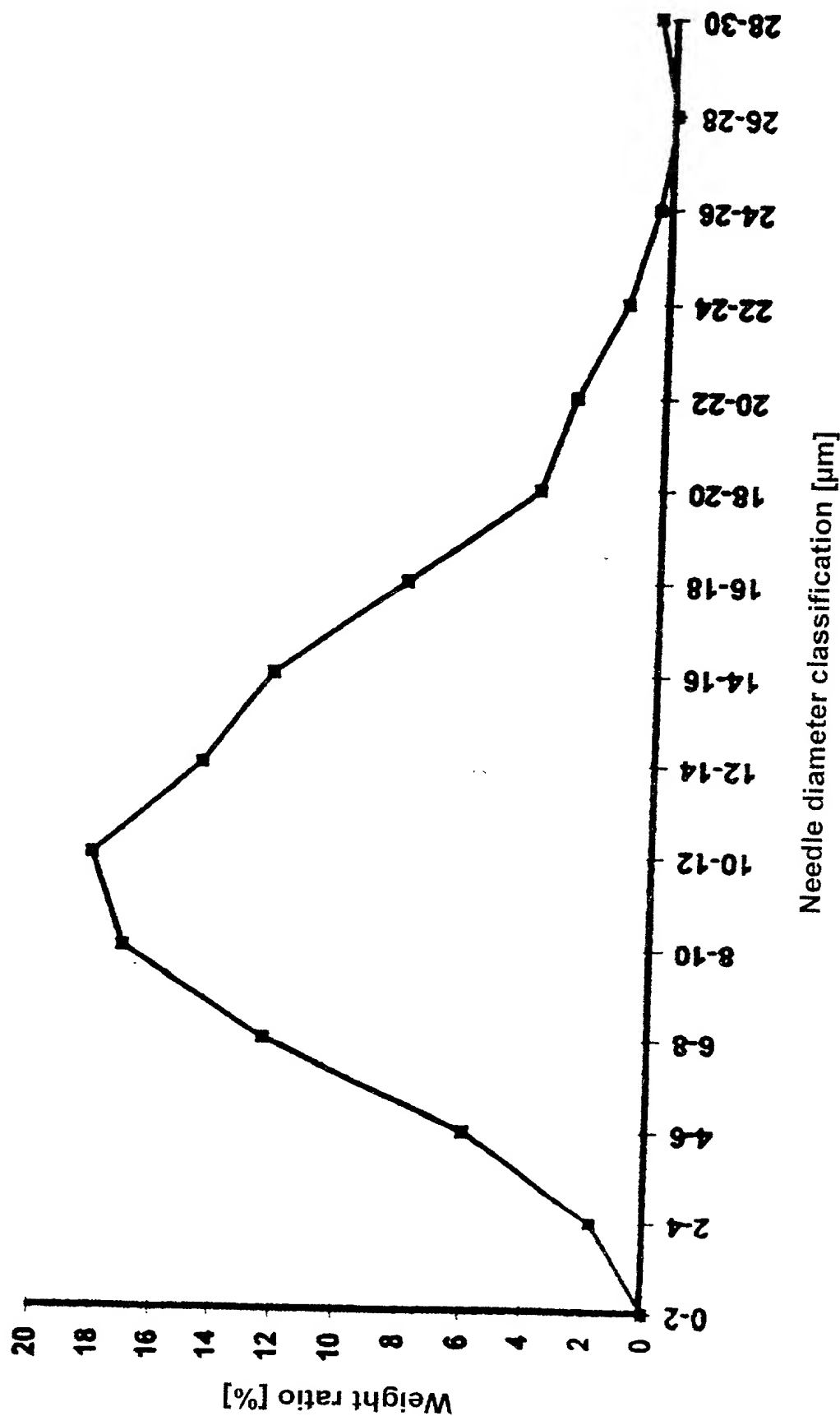
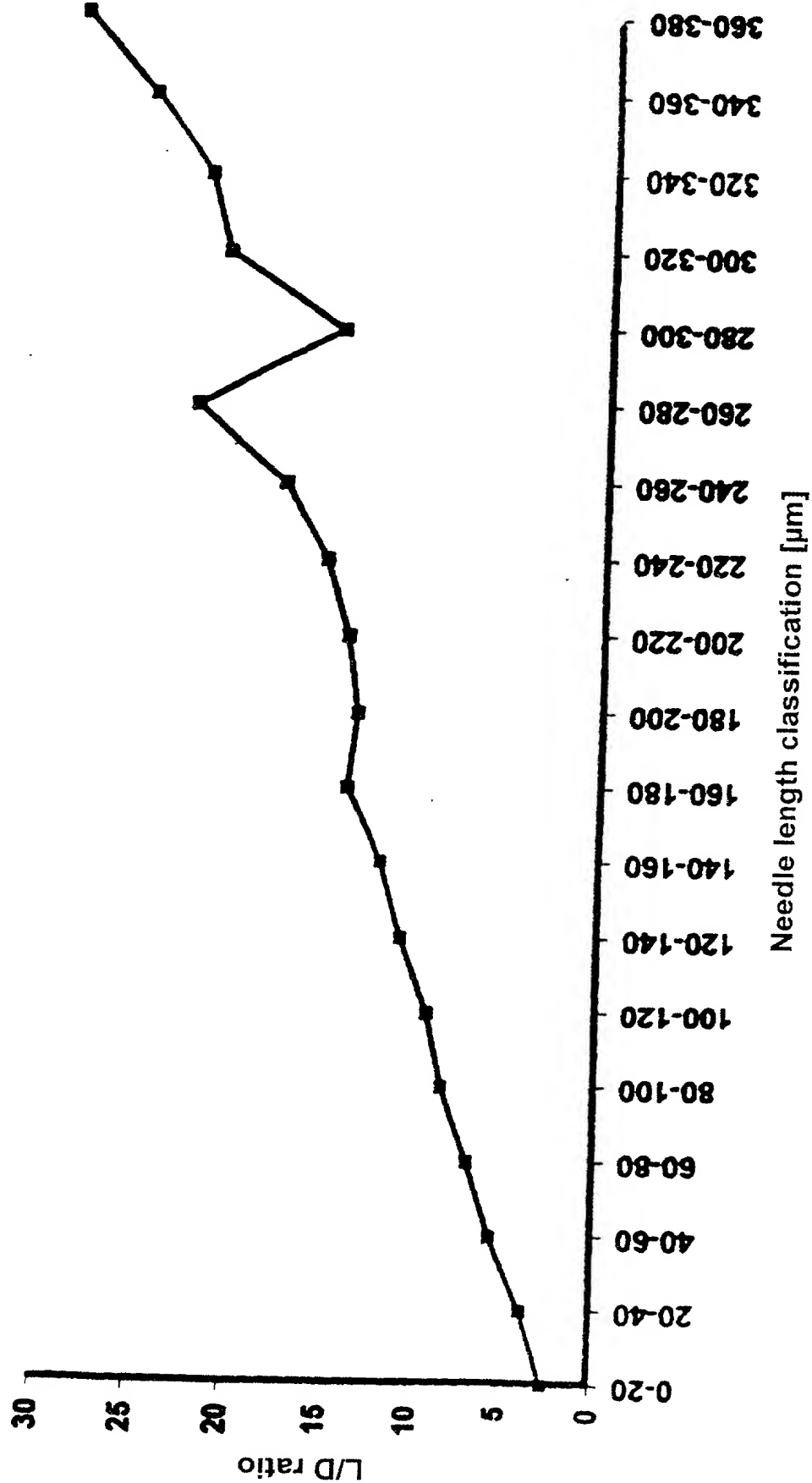


Figure 5

Average L/D ratio as a function of needle length



(Foreign associate use only)

## DECLARATION AND POWER OF ATTORNEY

Attorney's Docket No. 18584-0002

As a below named inventor, I hereby declare that:

My residence, post office address, and citizenship are as stated below next to my name. I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled: Method of Reducing the Dust Generation of Silicates, the specification of which

☒ is attached hereto.

☐ was filed on \_\_\_\_\_ as a PCT International Application No. \_\_\_\_\_ and was amended (if applicable) on \_\_\_\_\_.

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above. I do not know and do not believe that the same was ever known or used by others in the United States of America before my or our invention thereof, or patented or described in any printed publication in any country before my or our invention thereof or more than one year prior to the date of this application. I further state that the invention was not in public use or on sale in the United States of America more than one year prior to the date of this application. I understand that I have a duty of candor and good faith toward the Patent and Trademark Office, and I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, §1.56.

I hereby claim foreign priority benefits under Title 35, United States Code §119(a)-(d) or §365(b) of any foreign application(s) for patent or inventor's certificate, or §365(a) of any PCT international application which designated at least one country other than the United States of America, listed below and have also identified below any foreign application for patent or inventor's certificate disclosing subject matter in common with the above-identified specification and having a filing date before that of the application on which priority is claimed:

Country	App. No.	Date of Filing	Priority Claimed Under 35 USC §119
Germany	199 13 810.9	March 26, 1999	Yes <input checked="" type="checkbox"/> No <input type="checkbox"/>

I hereby claim the benefit under Title 35, United States Code, § 120 of any prior United States application(s), or §365(c) of any PCT international application designating the United States of America, listed below and, insofar as the subject matter of each claim of the present application is not disclosed in the prior United States or PCT international application in the manner provided by the first paragraph of Title 35, United States Code §112, I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations §1.56, which became available between the filing date of the prior application and the national or PCT international filing date of this application:

Application No.	Filing Date	Status: patented, pending, abandoned
PCT/EP00/1616	February 26, 2000	Pending

I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patents issuing thereon.

I hereby authorize the U.S. attorneys named herein to accept and follow instructions from **Sternagel Fleischer Godemeyer**, as to any action to be taken in the Patent and Trademark Office regarding this application, without direct communication between the U.S. attorney and the undersigned. In the event of a change in the persons from whom instructions may be taken, the U.S. attorney named herein will be notified by the undersigned.

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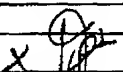
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Attorney Docket No.: 18584-0002

Title: Method of Reducing the Dust Generation of Silicates

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2.6

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